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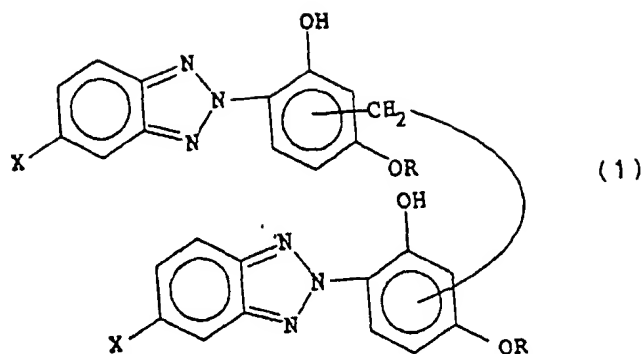
(54) [Title of the Invention] BENZOTRIAZOLE TYPE COMPOUNDS AND LIGHT STABILIZERS

(57) [Abstract]

[Object] To be able to improve the weather resistance and color fastness of organic materials.

[Means for Accomplishing Object] Light stabilizers containing a benzotriazole type compound that can be represented by the formula (1) given below.

[Chemical Formula 1]

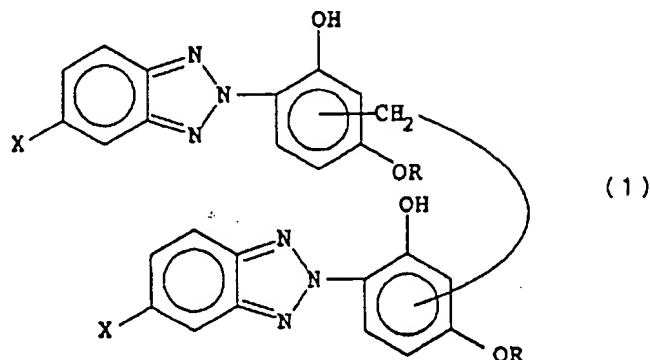


(in which X denotes a hydrogen atom or a chlorine atom, R denotes an alkyl group, an aryl group or an alkyl aryl group having the number of carbon atoms in the range of 1-18. The methylene group is bonded to the ortho position or the para position with respect to the hydroxyl group).

[Scope of the Patent Claims]

[Claim 1] Benzotriazole type compounds that can be represented by the formula (1) given below.

[Chemical Formula 1]



(in which X denotes a hydrogen atom or a chlorine atom, R denotes an alkyl group, an aryl group or an alkyl aryl group having the number of carbon atoms in the range of 1-18. The methylene group is bonded to the ortho position or the para position with respect to the hydroxyl group.)

[Claim 2] Light stabilizers containing the benzotriazole type compound in accordance with claim 1.

[Claim 3] Organic materials containing as a light stabilizer the benzotriazole type compound in accordance with claim 1.

[Detailed Description of the Invention]

[0001]

[Field of Industrial Application] The present invention pertains to new benzotriazole type compounds. More specifically, the present invention pertains to new benzotriazole type compounds that are useful as a light stabilizer and also pertains to light stabilizers.

[0002]

[Prior Art] It is well known that the conventional organic materials such as natural polymers, synthetic polymers, fats and lipids, etc., undergo deterioration due to the action of light and subsequently their mechanical strength is markedly reduced. Softening, becoming brittle and discoloration accompany the decrease in the mechanical strength. In order to prevent the light-induced deterioration of organic materials, various light stabilizers have been used and added during the synthesis and processing of the organic materials. Such light stabilizers include, e.g., compounds of benzophenone type, benzotriazole type, benzoate type, salicylate type, triazine type, hindered amine type, etc. It has been shown that these compounds show a stabilizing effect when used individually or in combination of a multiple type.

[0003]

[Object of the Invention] The prior-art light stabilizers are still not satisfactory in view of the possibility that they themselves may discolor, evaporate or bleed and from the standpoints of the weather resistance and the color change resistance of the organic materials to which these light stabilizers are added. Especially, when the processing temperature is increased, evaporation of a low-molecular-weight type light stabilizer will become vigorous and problems associated with evaporation-induced pollution of the work environment or clogging of the

facility may occur. Therefore, specific bisbenzotriazolyl phenol type compounds have been developed and described in Patent Disclosure No. Sho 61-[1986]-118,373 and Patent Disclosure No. Sho 61-[1986]-118,374. In Patent Disclosure No. Hei 10-[1998]-175,963, benzotriazole alkylene bisphenols were described. These compounds were developed in order to suppress evaporation by increasing the molecular weight through dimerization or addition of an alkyl phenol.

[0004] In addition, a specific bisbenzotriazole phenol was described in Patent Disclosure No. Hei 9-[1997]-136,060. It has been reported that this bisbenzotriazole phenol can undergo copolymerization with monomers capable of polymerization and condensation. Therefore, it can be introduced into the main chain or side chain of polycarbonate, polyester, polyurethane, polyurethane urea, etc., to impart long-term weather resistance to the products.

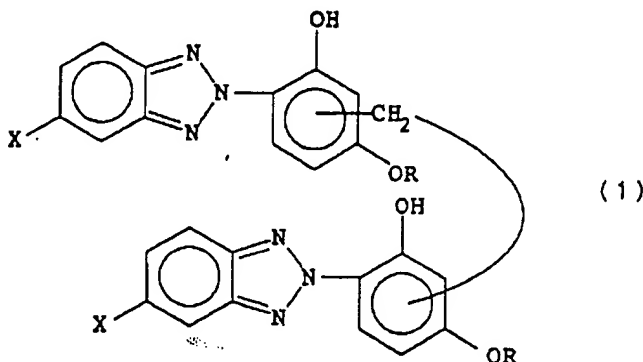
[0005] However, even these light stabilizers are still not satisfactory in view of the coloration of the light stabilizers themselves and from the standpoints of the weather resistance and color change resistance of the organic materials to which these light stabilizers are added.

[0006] Therefore, the object of the present invention is to provide non-coloring light stabilizers that can promote weather resistance and color change resistance. Another object of the present invention is to provide light stabilizers with the said characteristics that can also suppress or prevent evaporation or bleeding. Still another object of the present invention is to provide new benzotriazole type compounds.

[0007]

[Means for Accomplishing the Object] We carried out extensive studies and found that the new compounds mentioned above were the benzotriazole type compounds with increased molecular weight. Namely, the new compounds of the present invention are the benzotriazole type compounds that can be represented by the formula (1) given below.

[Chemical Formula 2]



(in which X denotes a hydrogen atom or a chlorine atom, R denotes an alkyl group, an aryl group or an alkyl aryl group having the number of carbon atoms in the range of 1-18. The methylene group is bonded to the ortho position or the para position with respect to the hydroxyl group.)

[0008] We also carried out extensive studies in order to overcome the problematic points associated with the prior-art light stabilizers and found that addition of specific benzotriazole type compounds with increased molecular weight to an organic material could improve its weather resistance and could also improve the extent of coloration before and after light irradiation. Namely, we discovered that both the weather resistance and the color change resistance of an organic material could be maintained at the same time. This discovery led us to develop the present invention. Namely, the present invention pertains to light stabilizers containing the benzotriazole type compounds that can be represented by the said formula (1).

[0009] When the benzotriazole type compounds that can be represented by the said formula (1) are used as a light stabilizer, the weather resistance and the color change resistance can be improved at the same time. In addition,

almost no coloration due to the light stabilizer itself will occur. Furthermore, synthesis and purification of the light stabilizers of the present invention with the said excellent characteristics are easy and they can be handled easily. Therefore, they can be used in the same way as the prior-art light stabilizers. Moreover, because of the high molecular weights, their evaporation can be suppressed or prevented and almost no bleeding will occur.

[0010] In addition, the present invention also pertains to organic materials that contain the benzotriazole type compounds of the said formula (1) as a light stabilizer.

- 10 [0011] When an organic material is allowed to contain the benzotriazole type compounds of the said formula (1) as a light stabilizer, the weather resistance of the organic material can be improved and at the same time discoloration of the organic material can be suppressed or prevented for a long time.

[0012]

[Embodiment of Implementation of the Invention] The benzotriazole type compounds of the present invention can be represented by the said formula (1). Namely, they are benzotriazole type compounds with increased molecular weight.

- 20 [0013] The alkyl groups in the said formula (1) with the number of carbon atoms in the range of 1-18 include, e.g., methyl group, ethyl group, propyl group, isopropyl group, butyl group, *s*-butyl group, *t*-butyl group, pentyl group, hexyl group, heptyl group, octyl group, nonyl group, decyl group, undecyl group, dodecyl group, tridecyl group, tetradecyl group, hexadecyl group, octadecyl group, etc. The alkyl group can be of straight chain or branched chain.

[0014] The aryl groups of R include, e.g., a phenyl group and the alkyl aryl groups include, e.g., various alkyl aryl groups such as a tolyl group, etc.

- 30 [0015] In the said formula (1), the methylene group ($-\text{CH}_2-$) is bonded to the ortho position or the para position on the benzene ring with respect to the hydroxyl group which is bonded to the benzene ring. Therefore, the benzotriazole type compounds that can be represented by the said formula (1) can be presented in three types of isomers. Since a reactive position is located in two positions, the ortho position and the para position with respect to the hydroxyl group, such polymers as a trimer, a tetramer, etc., may be formed.

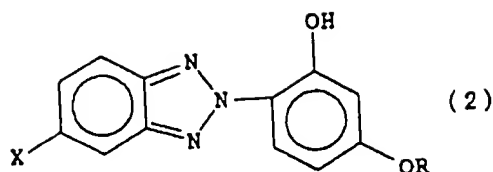
- 40 [0016] The three types of isomers include (1) a dimer in which the methylene group in one of the benzene rings with a bonded hydroxyl group is bonded to the ortho position with respect to the hydroxyl group while the methylene group on the other benzene ring with a bonded hydroxyl group is bonded to the ortho position with respect to the hydroxyl group, (2) a dimer in which the methylene group in one of the benzene rings with a bonded hydroxyl group is bonded to the ortho position with respect to the hydroxyl group while the methylene group on the other benzene ring with a bonded hydroxyl group is bonded to the para position with respect to the hydroxyl group and (3) a dimer in which the methylene group in one of the benzene rings with a bonded hydroxyl group is bonded to the para position with respect to the hydroxyl group while the methylene group on the other benzene ring with a bonded hydroxyl group is bonded to the para position with respect to the hydroxyl group.

[0017] There is no specific restriction as to the proportion of the three isomers that can be present in the compounds of the present invention. Furthermore, polymers such as a trimer, a tetramer, etc., may be present.

- 50 [0018] The benzotriazole type compounds that can be represented by the said formula (1) can be synthesized by any one of the various prior-art well-known methods. E.g., a benzotriazole type compound that can be represented by the formula (2) given below is reacted with an amine compound (NHR^1R^2) and formaldehyde (HCHO) to obtain a Mannich base that can be represented by the formula (3) given below. The benzotriazole type compound that can be represented by the formula (2) given below is then reacted with the Mannich base that can be represented by the formula (3) given below to obtain the target compound.

[0019]

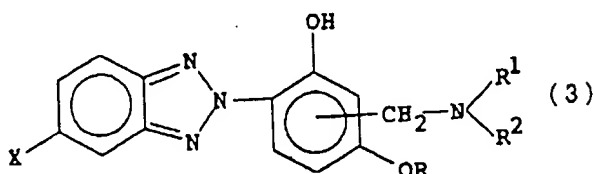
[Chemical Formula 3]



(in which X denotes a hydrogen atom or a chlorine atom, R denotes an alkyl group, an aryl group or an alkyl aryl group having the number of carbon atoms in the range of 1-18)

[0020]

[Chemical Formula 4]



(in which X denotes a hydrogen atom or a chlorine atom, R denotes an alkyl group, an aryl group or an alkyl aryl group having the number of carbon atoms in the range of 1-18, R¹ denotes a hydrogen atom or an alkyl group having the number of carbon atoms in the range of 1-6, R² denotes a hydrogen atom or an alkyl group having the number of carbon atoms in the range of 1-6. The amino methyl group that may have a substituting group is bonded to the ortho position or the para position with respect to the hydroxyl group).

[0021] This reaction may be carried out under an alkaline condition using an alkali. Proper alkalis that can be used in this reaction are hydroxides of alkali metal elements such as sodium hydroxide or hydroxides of alkali earth metal elements. In addition, this reaction can be carried out under a condition of normal pressure or reduced pressure and under a heated condition.

[0022] In the reaction between the benzotriazole type compound that can be represented by the said formula (2) and the Mannich base that can be represented by the said formula (3), the proportion of the dimers can be controlled by controlling the proportion of the two reacting compounds.

[0023] When the benzotriazole type compound that can be represented by the said formula (1) as a light stabilizer is blended into an organic material, the weather resistance of the organic material can be improved. In addition, the organic compound will almost not undergo discoloration for a long time and its color change resistance will be improved. Moreover, the benzotriazole type compound can be synthesized easily and its purification is also easy. It is also a compound that can be handled easily. Therefore, the benzotriazole type compounds of the present invention are useful as a stabilizer for suppressing or preventing light-induced deterioration of an organic material (a light stabilizer) and is especially useful as an ultraviolet light absorbing agent.

[0024] The organic materials of the present invention use the benzotriazole type compounds that can be represented by the said formula (1) as a light stabilizer. There is no specific restriction about these organic materials. Such organic materials include organic materials containing organic polymers such as plastics, resins, rubbers, etc. Various resins such as heat plasticizable resins, heat curable resins, ultraviolet light curable resins, etc., can be used as the organic polymers mentioned above. The organic materials can be used individually or in combination of more than two kinds.

[0025] Specifically, these organic materials include, e.g., such α -olefin type polymers as polyethylene, polypropylene, polybutene, polypentene, poly-3-methylbutylene, etc., such polyolefins as ethylene-vinyl acetate copolymer, ethylene-propylene copolymer, etc., such halogen containing synthetic resins as polyvinyl chloride, polyvinyl bromide, polyvinyl fluoride, polyethylene chloride, polypropylene chloride, polyethylene bromide, rubber chloride, vinyl chloride-vinyl acetate copolymer, vinyl chloride-ethylene copolymer, vinyl chloride-propylene copolymer, vinyl chloride-styrene copolymer, vinyl chloride-isobutylene copolymer, vinyl chloride-vinylidene chloride, vinyl chloride-ethylene-maleic anhydride ternary copolymer, vinyl chloride-styrene-acrylonitrile ternary copolymer, vinyl chloride-butadiene copolymer, vinyl chloride-isobutylene copolymer, vinyl chloride-propylene chloride copolymer, vinyl chloride-vinylidene chloride-vinyl acetate ternary copolymer, vinyl chloride-acrylic acid ester copolymer, vinyl chloride-maleic acid ester copolymer, vinyl chloride-methacrylic acid ester copolymer, vinyl chloride-acrylonitrile copolymer, internal plasticizable polyvinyl chloride, etc., petroleum resin, coumarone resins, copolymers of polystyrene or styrene and other copolymerizable monomers (e.g., maleic anhydride, butadiene, acrylonitrile, etc.), such styrene type resins as acrylonitrile-butadiene-styrene resin, acrylic acid ester-butadiene-styrene resin, methacrylic acid ester-butadiene-styrene resin, polyvinyl acetate, polyvinyl alcohol, polyvinyl formal, polyvinyl butyral, acryl resin, methacrylate resin, polyacrylonitrile, straight chain polyester, polyphenylene oxide, polyamide, polycarbonate, modified polyphenylene oxide, polyacetal, polyurethane, fibrin type resin, unsaturated polyester resin, phenol resin, urea resin, melamine resin, epoxy resin, silicone resin, polyethylene terephthalate, reinforced polyethylene terephthalate, polybutylene terephthalate, polysulfone type resin, polyether sulfone, polyphenylene sulfide, polyether ketone, polyether imide, polyoxybenzoyl, polyimide, polymaleimide, polyamide imide, etc. Moreover, rubbers can also be used as the organic materials. Such rubbers include, e.g., natural rubber, such synthetic rubbers as isoprene rubber, butadiene rubber, styrene-butadiene copolymerized rubber, acrylonitrile-butadiene copolymerized rubber, etc. Furthermore, they can be a blended product of these resins and rubbers, various cosmetic products and various photographic chemical reagents.

[0026] When these compounds are used as a light stabilizer for the organic material, it is desirable that the amount blended is in the range of 0.01-10 wt. % or preferably 0.05-5 wt. % of the total amount of the organic material used.

[0027] When the benzotriazole type compound of the present invention that can be represented by the said formula (1) is used as a light stabilizer, its concomitant use with other light stabilizer (e.g., other ultraviolet light absorbing agent, hindered amine type light stabilizer, etc.) or an antioxidant (e.g., a phenol type antioxidant, a sulfur type antioxidant, a phosphorus type antioxidant, etc.) can further promote the weather resistance, color change resistance, light resistance and heat stability.

[0028] There is no specific restriction as to other ultraviolet light absorbing agents that can be used together with the benzotriazole type compound that can be represented by the said formula (1). E.g., benzophenone type ultraviolet light absorbing agents, benzotriazole type ultraviolet light absorbing agents and other ultraviolet light absorbing agents may be used.

[0029] The benzophenone type ultraviolet light absorbing agents include, e.g., 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-methoxybenzophenone-5-sulfonic acid (anhydrous and trihydrate), 2-hydroxy-octyloxybenzophenone, 4-dodecyloxy-2-hydroxybenzophenone, 4-benzyloxy-2-hydroxybenzophenone, 2,2',4'-tetrahydroxybenzophenone, 2,2'-dihydroxy-4,4'-dimethoxybenzophenone, etc.

[0030] The benzotriazole type ultraviolet light absorbing agents include, e.g., 2-(2-hydroxy-5-methylphenyl)-2H-benzotriazole, 5-chloro-2-(3,5-di-*tert.*-butyl-2-hydroxyphenyl)chloro-2H-benzotriazole, 2-(3-*tert.*-butyl-2-hydroxy-5-methylphenyl)-5-chloro-2H-benzotriazole, 2-(3,5-di-*tert.*-pentyl-2-hydroxyphenyl)-2H-benzotriazole, 2-(3,5-di-*tert.*-butyl-2-hydroxyphenyl)-2H-benzotriazole, 2-(2H-benzotriazole-2-yl)-4-methyl-6-(3,4,5,6-tetrahydrophthalimidylmethyl)phenol, 2-(2H-benzotriazole-2-yl)-4-*tert.*-octyl-6-(3,4,5,6-tetrahydrophthalimidylmethyl)phenol, 2-(2-hydroxy-5-*tert.*-octylphenyl)-2H-benzotriazole, 2-(2-hydroxy-4-octyloxyphenyl)-2H-benzotriazole, 2-[2-hydroxy-3,5-bis(α,α -dimethylbenzyl)phenyl]-2H-benzotriazole, etc.

[0031] Other ultraviolet light absorbing agents include, e.g., phenyl salicylate, 4-*tert.*-butylphenyl salicylate, ethyl 2-cyano-3,3-diphenyl acrylate, 2'-ethylhexyl 2-cyano-3,3-diphenyl acrylate, 2'4'-di-*tert.*-butyl phenyl 3,5-di-*tert.*-butyl-4-hydroxy benzoate, 2-{4-[(2-hydroxy-3-alkyl)-oxy]-2-hydroxyphenyl}-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(4,6-diphenyl-1,3,5-triazine-2-yl)-5-(hexyl)oxyphenol, etc.

[0032] The hindered amine type light stabilizers include, e.g., bis(2,2,6,6-tetramethyl-4-piperidyl) sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)-2-(3,5-di-*tert.*-butyl-4-hydroxybenzyl)-2-butylmalonate, bis(1-acryloyl-2,2,6,6-tetramethyl-4-piperidyl)bis(3,-5-di-*tert.*-butyl-4-hydroxybenzyl)malonate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butene tetracarboxylate, poly{[6-(1,1,3,3-tetramethylbutyl)imino-*s*-triazine-2,4-diyl][(2,2,6,6-tetramethyl-4-piperidyl)imino]hexamethylene [(2,2,6,6-tetramethyl-4-piperidyl)imino]}, poly{(6-morpholino-*s*-triazine-2,4-diyl) [(2,2,6,6-tetramethyl-4-piperidyl)imino]hexamethylene [(2,2,6,6-tetramethyl-4-piperidyl)imino]}, 1-hydroxyethyl-2,2,6,6-tetramethyl-4-piperidinol/succinic acid condensation products, cyanyl chloride/*tert.*-octyl amine/1,6-bis(2,2,6,6-tetramethyl-4-piperidylamino)hexane condensation products.

- 10 [0033] Among the antioxidants, the phenol type antioxidants include, e.g., 2,6-*tert.*-butyl-4-methyl phenol, 4-hydroxymethyl-2,6-di-*tert.*-butyl phenol, 2,6-di-*tert.*-butyl-4-ethyl phenol, butylated hydroxy anisole, *n*-octadecyl 3-(4-hydroxy-3,5-di-*tert.*-butylphenyl)propionate, distearyl(4-hydroxy-3-methyl-5-*tert.*-butyl)benzyl malonate, tocopherol, 2,2'-methylene bis(4-methyl-6-*tert.*-butyl phenol), 2,2'-methylene bis(4-ethyl-6-*tert.*-butyl phenol), 4,4'-methylene bis(2,6-di-*tert.*-butyl phenol), 4,4'-butylidene bis(6-*tert.*-butyl-*m*-cresol), 4,4'-thio-bis(6-*tert.*-butyl-*m*-cresol), styrene transformed phenol, *N,N*-hexamethylene bis(3,5-di-*tert.*-butyl-4-hydroxyhydrocinnamide, calcium bis(3,5-di-*tert.*-butyl-4-hydroxybenzyl phosphonic acid ethyl ester), 1,1,3-tris(2-methyl-4-hydroxy-5-*tert.*-butyl phenyl) butane, 1,3,5-trimethyl-2,4,6-tris (3,5-di-*tert.*-butyl-4-hydroxybenzyl) benzene, tetrakis[3-(3,5-di-*tert.*-butyl-4-hydroxyphenyl)propionyloxymethyl] methane, 1,6-hexanediol bis[3-(3,5-di-*tert.*-butyl-4-hydroxyphenyl) propionate], 2,2-methylene bis(4-methyl-6-cyclohexyl phenol), 2,2'-methylene bis[6-(1-methylcyclohexyl)-*p*-cresol], 1,3,5-tris(4-*tert.*-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanuric acid, 1,3,5-tris(3,5-di-*tert.*-butyl-4-hydroxybenzyl) isocyanuric acid, triethylene glycol bis[3-(3-*tert.*-butyl-4-hydroxy-5-methylphenyl) propionate], 2,2'-oxamide bis[ethyl 3-(3,5-di-*tert.*-butyl-4-hydroxyphenyl) propionate], 6-(4-hydroxy-3,5-di-*tert.*-butyl anilino)-2,4-dioctyl thio-1,3,5-triazine, bis[2-*tert.*-butyl-4-methyl-6-(2-hydroxy-3-*tert.*-butyl-5-methylbenzyl)phenyl] terephthalate, 3,9-bis{2-(3-*tert.*-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl}-2,4,8,10-tetraoxaspiro[5.5]undecane, 3,9-bis{2-[3-(3,5-di-*tert.*-butyl-4-hydroxyphenyl)propionyloxy]-1,1-dimethylethyl}-2,4,8,10-tetraoxaspiro[5.5]undecane, etc.

- 30 [0034] The sulfur type antioxidants include, e.g., pentaerythritol tetrakis-(3-dodecylthiopropionate), didodecylthiodipropionate, ditetradecylthiodipropionate, dioctadecylthiodipropionate, etc.

[0035] The phosphoric acid type antioxidants include, e.g., tris(2,4-di-*tert.*-butylphenyl) phosphite, tetrakis(2,4-di-*tert.*-butylphenyl)-4,4'-biphenylene diphosphite, bis(2,4-di-*tert.*-butylphenyl)pentaerythritol diphosphonite, bis(2,6-di-*tert.*-butyl-4-methylphenyl)pentaerythritol diphosphite, 2,2'-methylene bis(4,6-di-*tert.*-butylphenyl)-2-ethylhexyl phosphite, tetrakis(2,4-di-*tert.*-butyl-5-methylphenyl)-4,4'-biphenylene diphosphonite, etc.

[0036] In addition, these compounds may be used concomitantly with a metal soap, a heavy metal inactivation agent, a nucleus forming agent, a plasticizer, a foaming agent, an antistatic agent, a flame retarder, a lubricant, a processing aide, etc.

- 40 [0037]

[Actual Examples] The present invention will be described in detail with the use of actual examples given below. However, the present invention is by no means limited to these actual examples.

[0038] 115.6 g of 6-(benzotriazole-2-yl)-3-*n*-octyloxy phenol, 50.0 g of dibutyl amine and 6.1 g of paraformaldehyde (purity: 92%) were placed in a 1,000-cc four-mouth flask. The mixture was refluxed at 95-98°C for about 6 hours and then heated to 120°C under a reduced pressure of 15 mmHg. 1.5 hours were needed to recover dibutyl amine and water. The reaction mixture was tested with the use of thin layer chromatography and the formation of two kinds of Mannich bases could be confirmed.

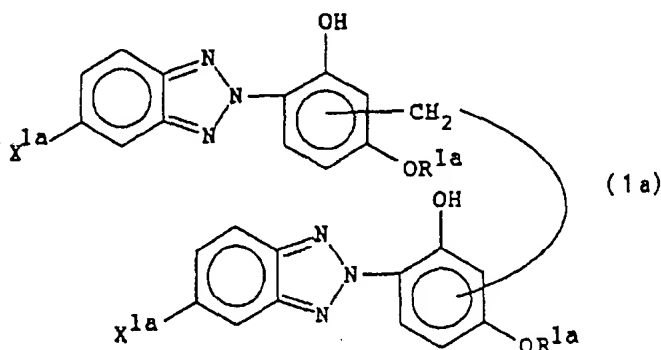
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[0039] Next, 5.0 g of an aqueous sodium hydroxide solution (sodium hydroxide concentration: 48 wt. %) were added. The reaction was completed in about 4 hours at about 160°C under a reduced pressure (15 mmHg). The reaction mixture was cooled to 120-130°C and was then extracted with 200 g of xylene and neutralized by adding 20.0 g of acetic acid. Addition of 200 mL of warm water to the extract resulted in deposition of crystals. After cooling to 5°C, the crystals were separated by filtration and washed with warm water and isopropyl alcohol to obtain 86.5 g of grayish white crystals (yield: 73.5%).

[0040] To 50.0 g of these crystals were added 50 mL of toluene, 150 mL of isopropyl alcohol and 1 g of activated charcoal and the mixture was refluxed. After filtration, the filtrate was cooled to 5°C and the deposited crystals were separated by filtration to obtain 48.6 g of slightly yellow granular crystals. The melting point of these crystals was in the range of 144-159°C. The crystals thus obtained were subjected to elementary analysis, mass spectral analysis and magnetic resonance spectral analysis and were confirmed to be the compound that could be represented by the formula (1a) given below. The results of these analyses are described below.

[0041]

[Chemical Formula 5]



(in which X^{1a} denotes a hydrogen atom, R^{1a} denotes an *n*-octyl group. The methylene group is bonded to the ortho position or the para position with respect to the hydroxyl group).

[0042] (Elementary analysis)

· C (carbon atom)	actually measured value: 71.3%	(calculated value: 71.3%)
· H (hydrogen atom)	actually measured value: 7.3%	(calculated value: 7.3%)
· N (nitrogen atom)	actually measured value: 12.3%	(calculated value: 12.2%)

(Nuclear magnetic resonance spectrum)

$^1\text{H-NMR}$ (in CDC, TMS standard)

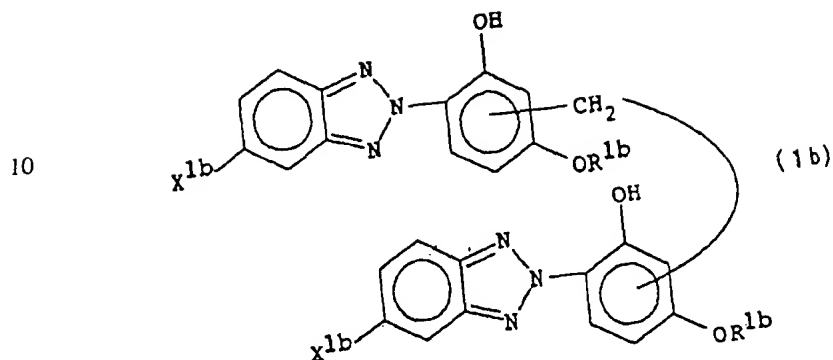
$\delta = 0.7536-1.9220$ (multiplet, $-(\text{CH}_2)_6\text{CH}_3$), $3.1858-4.4095$ (multiplet, $-\text{CH}_2-$, Ph-O-CH_2-), $6.5195-8.3087$ (multiplet, hydrogen atom of benzene ring), $11.3023-11.7012$ (multiplet, $-\text{OH}$)
(where Ph denotes a phenyl group).

[0043] These crystals were also analyzed by liquid chromatography with a detection wavelength of 254 nm and a mobile phase of (acetonitrile:dichloromethane = 1:1). It could be confirmed that the main product was a mixture of compounds with three peaks. This observation reflects the fact that three isomers are present in the benzotriazole type compound that can be represented by the said formula (1a); they are a dimer in which the methyl group is bonded to the ortho position and the ortho position with respect to the hydroxyl group, a dimer in which the methyl group is bonded to the ortho position and the para position with respect to the hydroxyl group and a dimer in which the methyl group is bonded to the para position and the para position with respect to the hydroxyl group.

[0044] (Actual Example 2) The reaction as described in Actual Example 1 was carried out using 6-(benzotriazole-2-yl)-3-*n*-butyloxyphenol in place of 6-(benzotriazole-2-yl)-3-*n*-octyloxyphenol. Light yellow powder crystals with a melting point in the range of 163.8-168.4°C could be obtained with a yield of 84.2%. These crystals were analyzed by elementary analysis and mass spectral analysis and were confirmed to be the benzotriazole type compound that could be represented by the formula (1b) given below. The results of the analyses are described below.

[0045]

[Chemical Formula 6]



20 (in which X^{1b} denotes a hydrogen atom, R^{1b} denotes an *n*-butyl group. The methylene group is bonded to the ortho position or the para position with respect to the hydroxyl group).

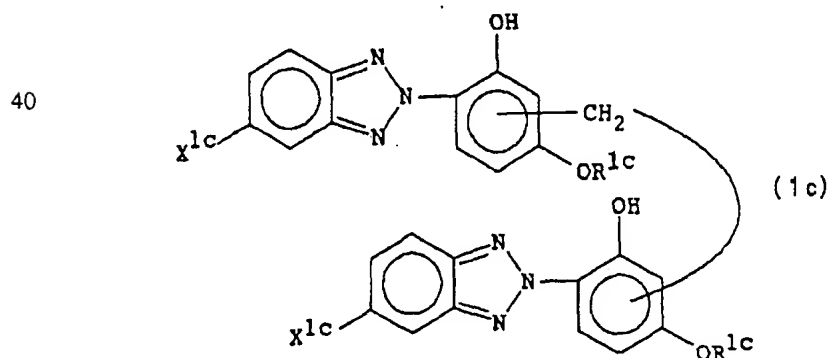
[0046] (Elementary analysis)

. C (carbon atom)	actually measured value: 68.6 %	(calculated value: 68.5 %)
. H (hydrogen atom)	actually measured value: 6.0 %	(calculated value: 5.9 %)
. N (nitrogen atom)	actually measured value: 14.4 %	(calculated value: 14.5 %)

30 [0047] (Actual Example 3) 6-(5-Chlorobenzotriazole-2-yl)-3-*n*-octyloxyphenol was used in place of 6-(benzotriazole-2-yl)-3-*n*-octyloxy phenol to carry out the reaction as described in Actual Example 1. Slightly green powder crystals with a melting point in the range of 130.0-150.0°C could be obtained with a yield of 62.9%. These crystals were analyzed by elementary analysis and mass spectral analysis and were confirmed to be the benzotriazole type compound that could be represented by the formula (1c) given below. The results of the analyses are described below.

[0048]

[Chemical Formula 7]



50 (in which X^{1c} denotes a hydrogen atom, R^{1c} denotes an *n*-octyl group. The methylene group is bonded to the ortho position or the para position with respect to the hydroxyl group).

[0049] (Elementary analysis)

. C (carbon atom)	actually measured value: 64.9 %	(calculated value: 64.8 %)
. H (hydrogen atom)	actually measured value: 6.4 %	(calculated value: 6.4 %)
. N (nitrogen atom)	actually measured value: 11.0 %	(calculated value: 11.1 %)
. Cl (chlorine atom)	actually measured value: 9.3 %	(calculated value: 9.3 %)

[0050] (Evaluation) The test compounds listed in Table 1, unstabilized polypropylene and calcium stearate were mixed for 5 minutes in a mixer using the blending composition or proportion given below. The mixture was then melted and kneaded at 180°C with the use of a mixing roll and the compound thus obtained was pressed in a heat press at 210°C to obtain a film with a thickness of 0.1 mm. This film was irradiated with light in the Carbon Arc Sunshine Weather Meter (63°C, 18 minutes/120 minutes with rain) and the infrared absorption spectrum was measured every 60 minutes. From the infrared absorption spectrum, the absorbance of methylene group (ABS 1) at 2740 cm⁻¹ and the absorbance of carbonyl group (ABS 2) at 1705-1720 m⁻¹ were determined. The time needed for the ABS 2/ABS 1 to become greater than 1 (induction period) was determined for evaluation of the weather resistance and light resistance. The results obtained are given under the column of "induction period (hour)" in Table 1. Of course, the longer the induction period, the better the weather resistance.

(Blending composition or proportion)

unstabilized polypropylene	:	100 weight parts
calcium stearate	:	0.1 weight part
test compound	:	0.2 weight part

[0051]

[Table 1]

No.		Test compound	Induction period (hours)
1	Example according to the present invention	Compound obtained in Actual Example 1	1320
2		Compound obtained in Actual Example 2	1260
3		Compound obtained in Actual Example 3	1320
4	Comparison Example	UVA-1	1140
5		UVA-2	1080
6		UVA-3	1140
7		No addition	120

UVA-1: 2-(3-*tert.*-butyl-2-hydroxy-5-methylphenyl)-5-chloro-2H-benzotriazole

UVA-2: 2-hydroxy-4-octyloxybenzophenone

UVA-3: 2,2'-methylenebis[(6-benzotriazole-2-yl)-4-*tert.*-octyl]phenol

[0052] (Light resistance and heat-induced color change tests) The test compounds listed in Table 2, unstabilized polypropylene and calcium stearate and tetrakis[3-(3,5-*tert.*-butyl-4-hydroxyphenyl)propionyloxymethyl]methane with the blending composition or proportion given below were granulated by extruding from a 40-mm-diameter extrusion granulator. A 5.5-ounce injection molding machine (a product of Toshiba, IS100E model) was then used to obtain a test piece. The test piece measuring 40 x 60 x 1 mm was irradiated with light in the Carbon Arc Sunshine Weather Meter (63°C, 18 minutes/120 minutes with rain) and the surface condition of the test piece was examined every 60 minutes. The time needed to form a crack on the surface of the test piece was measured. The results of the evaluation are given under the "weather resistance column" in Table 2. Separately, the test piece measuring 40 x 40 x 1 mm was installed and heated in a 160°C Gear Oven and the color difference ΔYI of the test piece before deterioration and 3 days after the start of the test was measured to evaluate the extent of heat induced color change. The results of this evaluation are given under the "heat induced color change" column in Table 2.

(Blending composition and proportion)

unstabilized polypropylene:	100 weight parts
calcium stearate:	0.1 weight part

tetrakis[3-(3,5-di-*tert.*-butyl-4-hydroxyphenyl)propionyloxymethyl]methane:
test compound:

0.1 weight part
0.2 weight part

[0053]

[Table 2]

No.		Test compound	Time needed to form surface crack (hr)	Heat-induced color change (ΔYI)
1	Example according to the present invention	Compound obtained in Actual Example 1	960	24.6
2		Compound obtained in Actual Example 2	900	25.3
3		Compound obtained in Actual Example 3	960	24.9
4	Comparison Example	UVA-1	780	35.6
5		UVA-2	660	39.8
6		UVA-3	660	35.2
7		No addition	120	24.0

UVA-1: 2-(3-*tert.*-butyl-2-hydroxy-5-methylphenyl)-5-chloro-2H-benzotriazole

UVA-2: 2-hydroxy-4-octyloxybenzophenone

UVA-3: 2,2'-methylenebis[(6-benzotriazole-2-yl)-4-*tert.*-octyl]phenol

[0054] (Color change resistance test) The test compound listed in Table 3, polyvinyl chloride, dioctyl phthalate, epoxidized soybean oil, calcium stearate and zinc stearate with the blending composition or proportion given below were mixed and kneaded at 180°C with the use of a mixing roll to obtain a film with a thickness of 0.2 mm. This film was irradiated with light in the Carbon Arc Sunshine Weather Meter (63°C, 18 minutes/120 minutes with rain) for 360 hours to examine the extent of color change. The results obtained are given in the "extent of color change" column in Table 3.

(Blending composition or proportion)

polyvinyl chloride:	100 weight parts
dioctyl phthalate:	48 weight parts
epoxidized soybean oil:	2 weight parts
calcium stearate:	1 weight part
zinc stearate:	0.1 weight part
test compound:	0.2 weight part

[0055]

[Table 3]

No.		Test compound	Extent of color change
1	Example according to the present invention	Compound obtained in Actual Example 1	Light yellow
2		Compound obtained in Actual Example 2	Light yellow
3		Compound obtained in Actual Example 3	Light yellow
4	Comparison Example	UVA-1	Yellowish brown
5		UVA-2	Brown
6		UVA-3	Yellowish brown
7		No addition	Dark brown

UVA-1: 2-(3-*tert.*-butyl-2-hydroxy-5-methylphenyl)-5-chloro-2H-benzotriazole

UVA-2: 2-hydroxy-4-octyloxybenzophenone

UVA-3: 2,2'-methylenebis[(6-benzotriazole-2-yl)-4-*tert.*-octyl]phenol

[0056] In Tables 1-3, the compound obtained in Actual Example 1 denotes the benzotriazole type compound that can be represented by the said formula (1a), the compound obtained in Actual Example 2 denotes the benzotriazole compound that can be represented by the said formula (1b) and the compound obtained in Actual Example 3 denotes the benzotriazole compound that can be represented by the said formula (1c). Moreover, UVA-1 denotes 2-(3-*tert.*-butyl-2-hydroxy-5-methylphenyl)-5-chloro-2H-benzotriazole, UVA-2 denotes 2-hydroxy-4-octyloxybenzophenone and UVA-3 denotes 2,2'-methylenebis[(6-benzotriazole-2-yl)-4-*tert.*-octyl]phenol. "No addition" means that the test compound was not used.

[0057] It can be seen from Table 1 that, compared with the conventional ultraviolet absorbing agents (namely, UVA-1, UVA-2 and UVA-3), the use of the compounds obtained in Actual Examples 1-3 (namely, benzotriazole type compounds with increased molecular weight) resulted in a prolonged induction period and excellent weather resistance.

[0058] It can be seen from Table 2 that, compared with the prior-art ultraviolet absorbing agents (namely, UVA-1, UVA-2 and UVA-3), the use of the compounds obtained in Actual Examples 1-3 (namely, benzotriazole type compounds with increased molecular weight) resulted in prolongation of the time needed for occurrence of surface crack and excellent light resistance. In addition, the extent of color change was very small and was about the same as that observed in the case of "no addition", indicating the excellence of the heat induced color change resistance of the ultraviolet light absorbing agents obtained in actual examples (compounds obtained in Actual Examples 1-3).

[0059] It can be seen from Table 3 that, compared with the prior-art ultraviolet absorbing agents (namely, UVA-1, UVA-2 and UVA-3), the use of the compounds obtained in Actual Examples 1-3 (namely, benzotriazole type compounds with increased molecular weight) resulted in a decrease in the extent of color change and improved color change resistance.

[0060] Therefore, the use of the compounds of the present invention, namely, the benzotriazole type compounds that can be represented by the said formula (1) can improve the weather resistance and the color change resistance of an organic material at the same time.

[0061]

[Effect of the Invention] The compounds of the present invention can improve the weather resistance and color change resistance of an organic material and can suppress or prevent color change of the organic material for a prolonged period of time.